DIETARY EXPOSURE TO DIOXINS IN JAPAN: NATIONWIDE TOTAL DIET STUDY 2002-2006

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Introduction

Food is generally recognized as the main route of the human intake of polychlorinated dibenzo-*p*-dioxins (PCDDs), dibenzofuranes (PCDFs) and dioxin-like polychlorinated biphenyls (dl-PCBs), which are known collectively as dioxins. It is therefore important to estimate the dietary intake of dioxins for risk assessments. A total diet study (TDS), also known as a market basket study, is a useful method for estimating the average dietary intake of contaminants. Therefore, we have been performing a nationwide TDS of dioxins in Japan annually since 1998. Here, we report the TDS result for fiscal year 2006 and also discuss the time trend of the dietary intake of dioxins from TDS results obtained over the last 5 years (fiscal years 2002-2006).

Materials and Methods

Sample preparations: TDS samples were prepared in 7 regions (Hokkaido, Tohoku, Kanto, Chubu, Kansai, Chugoku-Shikoku and Kyushu) across Japan. This involved 9 locations in fiscal year 2006 (12 locations in fiscal year 2002, 11 locations in fiscal year 2003 and 9 locations in fiscal years 2004 and 2005). More than one hundred food items were collected from supermarkets in each location. The TDS samples in these locations were designed based on official food classification and consumption data obtained by the National Health and Nutrition Survey in Japan. These food samples were cooked or prepared for consumption in typical ways. These samples were then blended to form 14 food group composites. Table 1 shows the classification of the 14 food groups and their daily consumption in Kanto, one of the 7 regions. The consumption pattern does not differ significantly from the consumption patterns in the other regions. We prepared three samples (shown as A, B and C in Table 1) in food groups 10–12. As far as possible, we chose different food items, such as different fish species and different edible parts of meat. This was because food items classified into these groups have a relatively wide range of dioxin concentrations[1]. Therefore, the choice of food items has a great influence on the estimated dioxin intake. With the remaining food groups (1–9, 13, and 14), the 7 regions were classified into five blocks, and composite food groups in each block were prepared by mixing regional samples according to regional consumption.

Dioxin analyses: The dioxins were extracted, purified and analysed according to the guideline for the analysis of dioxins in foods in Japan. In brief, all the samples were spiked with $^{13}C_{12}$-labelled internal quantification standards. Food groups 1, 2 and. 5–8 (50 g) were twice extracted by shaking them with 50% acetone/*n*-hexane. The extracts were treated with concentrated sulphuric acid, and then purified on a silver nitrate/silica gel column, followed by an alumina column. The mono-*ortho* PCBs fraction and non-*ortho* PCB and PCDD/Fs fraction were separated on the alumina column. The former fraction was further purified by DMSO partition followed by a silica gel column. The latter fraction was further purified on an activated carbon-dispersed column. Both fractions were spiked with $^{13}C_{12}$-labelled recovery standards, and subjected to HRGC/HRMS equipped with a solvent-cut system with a large volume injector. Food groups 3, 4 and 9–13 (10–50 g) were digested with aqueous potassium hydroxide at room temperature. The alkaline hydrolysates were extracted with *n*-hexane. The extracts were treated with concentrated sulphuric acid, and then purified on a nitrate/silica gel column, followed by an activated carbon-dispersed column. The mono-*ortho* PCBs fraction and the non-*ortho* PCBs and PCDD/Fs fraction were separated on the activated carbon-dispersed column. The former fraction was further purified by DMSO partition followed by a silica gel column. The latter fraction was further purified on an alumina column. Both the fractions were spiked with $^{13}C_{12}$-labelled recovery standards, and subjected to HRGC/HRMS. Food group 14 (5 L) was filtered through a C18 disk, and then the disk was subjected to Soxhlet extraction with acetone followed by toluene. The extract was treated with concentrated sulphuric acid, and then purified on a nitrate/silica gel column, followed by an activated carbon-dispersed column. The fraction was spiked with $^{13}C_{12}$-labelled recovery standards, and subjected to HRGC/HRMS. The PCDD/Fs and non-*ortho* PCBs were determined using an SP-2331 and DB-17 column. The mono-*ortho* PCBs were determined using an HT8-PCB column. The limits of detection (LODs) for PCDD/Fs were as follows: 0.01–0.05 pg/g in food groups 1–3 and 5–13; 0.05–0.2 pg/g in food group 4; 0.1–0.5 pg/L in food group 14. The limits of detection for dl-PCBs were as follows: 0.1–1 pg/g in food groups 1–3 and 5–13; 0.05–0.2 pg/g in food group 4; 1–10 pg/L in food group 14. The TEQ concentrations were calculated using WHO-TEFs (1998). The total TEQ in a sample was calculated assuming that all isomer concentrations lower than the LODs were equal to zero (ND=0) or half of the LODs (ND=1/2LOD).

Results and Discussion

Table 2 summarizes the daily dietary intakes of dioxins in the 7 regions in fiscal year 2006. The national mean intake calculated at ND=0 was 52.2 pg-TEQ/day, corresponding to 1.04 pg-TEQ/kg/day for an adult weighing 50 kg. The intake was about one fourth of the tolerable daily intake (TDI) of 4 pg-TEQ/kg/day set by the Japanese government in 1999[2]. The intakes in the 7 regions ranged from 0.38 to 1.94 pg-TEQ/kg/day. The maximum intake was still about half of the TDI. As the result of analysing three samples in each of the food groups 10–12, we found that there were 1.5– to 4.5–fold differences between the minimum and maximum intakes of dioxins in each region (shown as columns #1 and #3 in Table 2). This is mainly because the three samples of food group 10 (fish and shellfish) had a relatively wide range of dioxin concentrations owing to the different varieties of fish species used to prepare them. The dietary intakes calculated at ND=1/2LOD are also given as a reference. The national mean intake was 111.9 pg-TEQ/day (2.24 pg-TEQ/kg/day), which is about twice the mean intake calculate at ND=0. Recently, Sasamoto *et al.* carried out TDS in metropolitan Tokyo, Japan[3], and they reported that the daily dioxin intake was 1.55 pg-TEQ/kg/day at ND=0 (2.20 pg-TEQ/kg/day at ND=1/2LOD) in 2004. Their result is within the range of dietary intakes of dioxins obtained from the 7 regions in our study.

Table 3 shows the mean intakes of dioxins from each food group in fiscal year 2006. The dioxin intakes were highest from fish and shellfish (group 10) followed by meat and eggs (group 11), and milk and dairy products (group 12) at ND=0, and were highest from fish and shellfish (group 10) followed by beverages (group 9), and rice and rice products (group 1) at ND=1/2LOD. The TEQ contributions of the fish and shellfish group were noticeable in the total TEQs (about 90% at ND=0 and 43% at ND=1/2LOD). A much greater difference of intake was observed between the estimates obtained at ND=0 and ND=1/2LOD for beverages, and rice and rice products, because these food groups contain high percentages of non-detected data as well as having high daily consumptions.

Figure 1 shows the dioxin intakes obtained from our TDS results between fiscal years 2002 and 2006. The national mean intakes were within a range of 1.0 to 1.5 pg-TEQ/kg/day at ND=0, which is well below the Japanese TDI. Additionally, the maximum dioxin intakes observed during the same period were below the TDI, although some maximum intakes were near the TDI. The latest mean intake was the lowest value in the last 5 years. The mean intakes appeared to be decreasing slowly, although the each TDS sample had a wide range of dioxin intakes.

TEF values have been revised recently. WHO advises that the TEF_{2005} be used to replace the previous TEF_{1998}. The recalculated average intake using TEF_{2005} in fiscal year 2006 showed a decrease of about 15% in the intake calculated with TEF_{1998} (Figure 2). This is mainly because the TEF_{2005} of PCB 118 (0.00003) is slightly lower than that of TEF_{1998} (0.0001).

Thus, the estimated dioxin intakes in the last 5 years were below the Japanese TDI. However, in some cases, the maximum intakes were near the TDI, and so continuous monitoring of the dietary intake of dioxins is recommended.

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References
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Table 1: Composition of 14 food groups in Kanto region (FY 2006)

No.	Food group[a]		No. of food stuffs	Daily consumption (g/day)
1	Rice and rice products		3	325.3
2	Cereals, seeds and potatoes		21	181.3
3	Sugars and confectioneries		9	35.7
4	Fats and oils		6	12.1
5	Pulses		7	55.3
6	Fruits		11	130
7	Green vegetables		8	106
8	Other vegetables, mushrooms and seaweed		21	206.7
9	Beverages		8	583.2
10	Fish and shellfish	A	26	85.8
		B	17	
		C	17	
11	Meat and eggs	A	12	116.1
		B	11	
		C	11	
12	Milk and dairy products	A	6	152.5
		B	6	
		C	6	
13	Other foods (seasoning)		16	94.2
14	Drinking water		1	600

[a] Three samples, shown as A, B and C, in food groups 10-12 were prepared using different food items as far as possible classified into each food group.

Table 2: Daily dietary intake of dioxins in Japan (FY 2006)

Region		Dioxin intake (pg-TEQ/day)[a]							
		ND=0				ND=1/2LOD			
		#1	#2	#3	Mean	#1	#2	#3	Mean
Hokkaido		18.9	22.3	85.4	42.2	78.2	81.9	140.7	100.3
Tohoku		26.3	53.1	92.5	57.3	83.6	108.0	145.4	112.3
Kanto Ⅰ		29.8	46.8	73.4	50.0	87.5	103.1	129.2	106.6
Ⅱ		39.4	50.1	69.0	52.8	97.6	107.9	126.0	110.5
Chubu Ⅰ		33.6	43.6	50.2	42.5	92.1	101.9	108.1	100.7
Ⅱ		23.2	35.1	62.1	40.1	84.6	95.9	122.0	100.8
Kansai		49.0	74.8	88.2	70.7	116.6	140.9	153.2	136.9
Chugoku-Shikoku		46.7	53.9	97.2	65.9	108.8	116.0	159.2	128.0
Kyushu		30.7	32.5	82.6	48.6	93.6	95.8	142.9	110.8
National mean	pg-TEQ/day	52.2 (18.9 - 97.2)				111.9 (78.2 - 159.2)			
(min - max)	pg-TEQ/kg/day[b]	1.04 (0.38 - 1.94)				2.24 (1.56 - 3.18)			

[a] Dioxin intakes in each region are shown in three different ways: #1, #2 and #3 were calculated by using minimum, median and maximum intakes, respectively, of food groups 10-12 in each region.

[b] Assuming an adult weight of 50 kg.

Table 3: Daily dietary intakes of dioxins from each food group (FY 2006)

No.	Food group	Dioxin intake (pg-TEQ/day)			
		ND=0		ND=LOD/2	
		Mean	Ratio (%)	Mean	Ratio (%)
1	Rice and rice products	< 0.1	< 0.1	13.0	11.6
2	Cereals, seeds and potatoes	0.3	0.5	7.1	6.3
3	Sugars and confectioneries	0.1	0.2	1.3	1.2
4	Fats and oils	0.1	0.1	1.5	1.3
5	Pulses	0.0	< 0.1	1.7	1.5
6	Fruits	< 0.1	< 0.1	3.4	3.1
7	Green vegetables	0.1	0.1	2.6	2.3
8	Other vegetables, mushrooms and seaweed	0.1	0.1	5.6	5.0
9	Beverages	< 0.1	< 0.1	14.4	12.8
10	Fish and shellfish	47.0	90.0	47.9	42.8
11	Meat and eggs	3.5	6.7	5.7	5.1
12	Milk and dairy products	1.1	2.0	4.9	4.3
13	Other foods (seasoning)	0.1	0.1	2.6	2.3
14	Drinking water	< 0.1	< 0.1	0.2	0.1
	Total	52.2	100.0	111.9	100.0

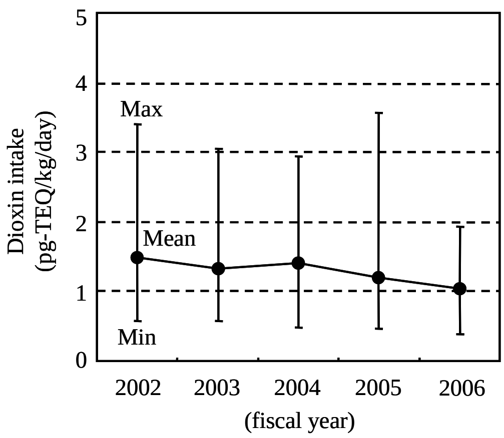

Figure 1: Time trend for dietary intake of dioxins (ND=0)

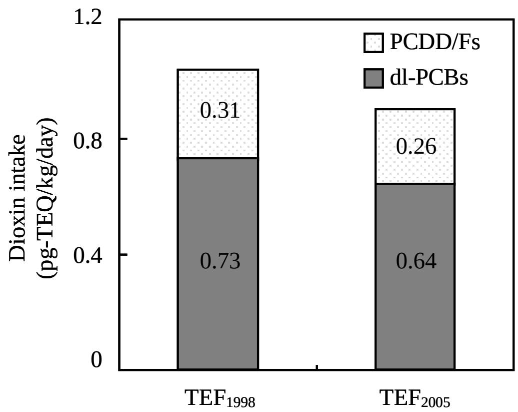

Figure 2: Comparison of dioxin intakes calculated using TEF_{1998} and TEF_{2005} (FY 2006)